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(54) Title of the invention New Polymerizable Unimer

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(72) Inventor Yoshida Haruo

Oita ken, Oita shi, Oji Nakanoshu, 2.

Showa Denko Kabushiki Kaisha Oita
Kenkyusho Nai

(Internal to Oita Research Center of Showa
Electrical Co. Ltd.)

(72) Inventor Tatsuke Hiroyuki

Oita ken, Oita shi, Oji Nakanoshu, 2.

Showa Denko Kabushiki Kaisha Oita
Kenkyusho Nai

(Internal to Oita Research Center of Showa
Electrical Co. Ltd.)

Abe Ichiro

Oita ken, Oita shi, Oji Nakanoshu, 2.

Showa Denko Kabushiki Kaisha Oita
Kenkyusho Nai

(Internal to Oita Research Center of Showa

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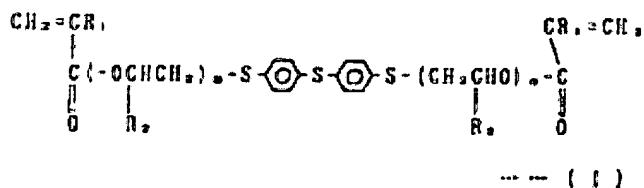
A Detailed Description

1. Title of the invention

New Polymerizable Unimer.

2. Scope of patent claims

4, 4' – bis [ω - (metha) acryloyl (polyoxy alkylene) thio] phenyl sulfide compound shown by the general formula (1) given below.



(Wherein, R_1 and R_2 independently show hydrogen or methyl radical and m and n are integers from 1 ~ 5.)

3. Detailed description of the invention

[Industrial applicability]

The present invention relates to a new di-functional polymerizable unimer that can be used in the manufacture of transparent hardened material having high heat resistance, low water absorbance and high refractive index. In further details, the present invention relates to 4, 4' – bis [ω - (metha) acryloyl (polyoxy alkylene) thio] phenyl sulfide compound that is suitable as raw

material of optical material, coating agent, injection molding material or light sensitive resin.

[Techniques of the past]

Polymers such as polystyrene group resins, poly methyl methacrylate group resins, polycarbonate resins, di ethylene glycol di allyl carbonate are used as organic optical material since past and their demand is rising due to their lightweight, safety, excellent operability and dyeing property.

However, the organic optical material of the past such as poly methyl methacrylate group resin, has high moisture absorbing property as the resin property and as a result of moisture absorption, its shape and refractive index changes and it becomes unstable as optical material. Moreover, polystyrene group resin and polycarbonate resin have the disadvantage of generation of optical birefringence (double refraction), scattered light, lowering of transparency due to temporal change etc. Furthermore, di ethylene glycol di allyl carbonate polymer has limited application scope as optical material due to low refractive index (refractive index = 1.499).

Different resins used for optical material have been proposed in order to reform these disadvantages. As examples, Patent number Sho 57 – 28115, Sho 57 – 28116, Sho 59 – 184210, Sho 60 – 7314, Sho 60 – 179406, Sho 60 – 217301, Sho 60 – 186514, Sho 60 – 166307, Sho – 103301, Sho 60 – 124607, Sho 62 – 232414, Sho 62 – 235901, Sho 62 – 267316, Sho 63 - 15811, Sho 63 – 46213, Sho 63 – 72707 etc. can be given.

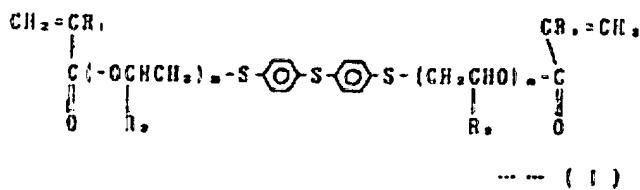
However, these composites obtained by the technique of past are optically uneven, under coloration, lack dimension stability due to which these cannot be used as optical material.

[Problems the invention solves]

The present invention aims at subjugating the problems of the resin used for optical material of the past and offers a new polymerizable unimer that can be used in the manufacture of hardened material having balance between properties such as optical evenness, low water absorbance, heat resistance, that can also be used as material of coating agent, sealing agent, paint, adhesive, injection molding material etc.

[Method to solve the problems]

The present invention offers the new polymerizable unimer that achieves the aim mentioned above. Namely, the present invention offers 4, 4' – bis [ω - (metha) acryloyl (polyoxy alkylene) thio] phenyl sulfide compound shown by the general formula (1) given below.

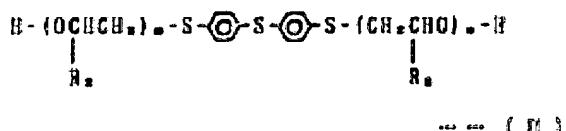


(Wherein, R₁ and R₂ independently show hydrogen or methyl radical and m and n are integers

from 1 ~ 5.)

The compound shown by the general formula (1) of the present invention can be synthesized by different methods and the concrete examples have been given below. Moreover, conditions of well-known reaction techniques can be used as the synthesis reaction conditions in this method.

(i) The method of carrying out esterification reaction by reacting divalent alcohol shown by the general formula (II) given below with (metha) acrylic acid (in the present invention, acrylic acid and / or methacrylic acid).



(Wherein, R_2 shows hydrogen or methyl radical and m and n are integers from 1 ~ 5.)

(ii) The method of carrying out esterification reaction between divalent alcohol shown by the general formula (II) given above and lower grade alkyl ester of (metha) acrylic acid.

(iii) The method of reacting divalent alcohol shown by the general formula (II) given above with salt of (metha) acrylic acid in the presence of base.

The divalent alcohol shown by the general formula (II) given above can be obtained by adding substituted or non-substituted alkylene oxide to 4, 4' – di mercapto phenyl sulfide or adding and condensing substituted or non-substituted alkylene carbonate to 4, 4' – di mercapto phenyl sulfide.

Therefore, in this method, value of m or n of the compound shown by the general formula (II) is decided by the reaction molar ratio of substituted or non-substituted alkylene oxide and 4, 4' – di mercapto phenyl sulfide or reaction molar ratio of substituted or non-substituted alkylene carbonate and 4, 4' – di mercapto phenyl sulfide.

As regards the substituted or non-substituted alkylene oxide used at the time of synthesizing polyoxy alkylene compound, ethylene oxide or propylene oxide can be used. Moreover, as regards the substituted or non-substituted alkylene carbonate, ethylene carbonate or propylene carbonate can be used.

4, 4' – bis [ω - (metha) acryloyl (polyoxy alkylene) thio] phenyl sulfide compound shown by the general formula (1) of the present invention is a colorless transparent compound that is liquid at normal temperature and has high refractive index and has high polymerization activity.

Therefore, this compound can be easily polymerized independently or by mixing with different polymerizable unimers possessing olefin type double bond, by using common radical polymerization initiator. Moreover, the obtained hardened material is colorless transparent and has high refractive index and moreover, has low water absorbance due to which it is suitable as optical material, paint, adhesive, coating agent, injection molding material etc.

The polymerizable unimer possessing olefin type double bond that is copolymerizable with the

compound shown by the general formula (1) can be uni-functional or multifunctional and it should be compatibility with 4, 4' – bis [ω - (metha) acryloyl (polyoxy alkylene) thio] phenyl sulfide compound. Moreover, polymerizable unimer possessing copolymerizable olefin type double bond can be as a mixture of 2 or more than 2 types and there is no particular restriction over the mixing proportion.

As regards the polymerizable unimer possessing olefin type double bond, unsaturated fatty acid ester, aromatic vinyl compound, unsaturated fatty acids and their derivatives, unsaturated dibasic acid and their derivatives, vinyl cyanide compounds such as (metha) acrylonitrile etc. can be given.

As regards the unsaturated fatty acid ester, alkyl (metha) acrylate such as methyl (metha) acrylate, ethyl (metha) acrylate, octyl (metha) acrylate, do decyl (metha) acrylate, octa decyl (metha) acrylate, cyclo hexyl (metha) acrylate, methyl cyclo hexyl (metha) acrylate, (iso) volnyl (?) (metha) acrylate, adamantlyl (metha) acrylate etc., acrylic acid aromatic ester such as phenyl (metha) acrylate, benzyl (metha) acrylate, 1 – naphthyl (metha) acrylate, fluoro phenyl (metha) acrylate, chloro phenyl (metha) acrylate, bromo phenyl (metha) acrylate, tri bromo phenyl (metha) acrylate, methoxy phenyl (metha) acrylate, cyano phenyl (metha) acrylate, bi phenyl (metha) acrylate, bromo benzyl (metha) acrylate etc., halo alkyl (metha) acrylate, fluoro methyl (metha) acrylate, chloro methyl (metha) acrylate, bromo ethyl (metha) acrylate, tri chloro methyl (metha) acrylate etc., 2 – hydroxy ethyl (metha) acrylate, (metha) acrylic acid polyethylene glycol ester etc., (metha) acrylic acid ester such as glycidyl (metha) acrylate, alkyl amino (metha) acrylate etc. can be given. Moreover, α - substituted acrylic acid ester such as α - fluoro acrylic acid ester, α - cyano acrylic acid ester etc. can be given.

As regards the aromatic vinyl compound, styrene, α - substituted styrene such as α - methyl styrene, α - ethyl styrene, α - chloro styrene etc., nuclear substituted styrene such as fluoro styrene, chloro styrene, bromo styrene, chloro methyl styrene, methoxy styrene etc. can be given.

As regards the unsaturated fatty acids and their derivatives, (metha) acrylamides such as (metha) acrylamide, N, N – di methyl (metha) acrylamide, N, N – di ethyl (metha) acrylamide etc., (metha) acrylic acid etc. can be given.

As regards the unsaturated dibasic acids and their derivatives, N – substituted maleimide such as N – methyl maleimide, N – ethyl maleimide, N – butyl maleimide, N – cyclo hexyl maleimide, N – phenyl maleimide, N – methyl phenyl maleimide, N – chloro phenyl maleimide, N – carboxyl phenyl maleimide etc., maleic acid, anhydrous maleic acid, fumaric acid etc. can be given.

As regards the polymerizable unimer possessing olefin type double bond that can be mixed with the compound shown by the general formula (1) other than uni-functional vinyl unimer mentioned above, bridging multifunctional monomer can be given. For example, di (metha)

acrylate such as ethylene glycol di (metha) acrylate, di ethylene glycol di (metha) acrylate, tri ethylene glycol di (metha) acrylate, tetra ethylene glycol di (metha) acrylate, tri propylene glycol di (metha) acrylate, 1, 3 – butylene glycol di (metha) acrylate, 1, 4 – butane diol di (metha) acrylate, 1, 5 – pentadiol di (metha) acrylate, 1, 6 – hexane diol di (metha) acrylate, neo pentyl glycol di (metha) acrylate, hydroxy pivalic acid neo pentyl glycol ester di (metha) acrylate, oligoester di (metha) acrylate, poly butadiene di (metha) acrylate, 2, 2 – bis (4 – (metha) acryloyloxy phenyl) propane, 2, 2 – bis (4 – (ω - (metha) acryloyloxy poly ethoxy)phenyl) propane, 2, 2 – bis (4 – (ω - (metha) acryloyloxy poly ethoxy) di bromo phenyl) propane, 2, 2 – bis (4 – (ω - (metha) acryloyloxy poly ethoxy)phenyl) propane, bis (4 – (ω - (metha) acryloyloxy poly ethoxy)phenyl) methane etc., bifunctional bridging monomer such as di allyl phthalate, di allyl iso phthalate, di allyl terephthalate, di vinyl benzene, N, N' – m – phenylene bis maleimide etc., tri-functional bridging monomer such as tri methylol ethane tri (metha) acrylate, tri methylol propane tri (metha) acrylate, penta erythritol tri (metha) acrylate, tri (metha) allyl is cyanulate, tri allyl tri meritate (?), di allyl chlorindate (?) etc., quadri-functional bridging monomer such as penta erythritol tetra (metha) acrylate etc. can be given.

The radical polymerization initiator that can generate radical by heat, microwaves, infrared rays or ultraviolet rays can be used.

As regards the radical polymerization initiator that can be used at the time of carrying out polymerization by heat, microwaves and infrared rays, azo group compound such as 2, 2' - azo bis iso butylonitrile, 2, 2' - azo bis iso valeronitrile (?) etc., ketone peroxide such as methyl ethyl ketone peroxide, methyl iso butyl ketone peroxide, cyclo hexanone peroxide, acetyl acetone peroxide etc., di acyl peroxide such as iso butyl peroxide, 2, 4 – di chloro benzoyl peroxide, o – methyl benzoyl peroxide, lauroyl peroxide, p – chloro benzoyl peroxide etc., hydro peroxide such as 2, 4, 4 – tri methyl pentyl – 2 – hydro peroxide, di iso propyl benzene peroxide, cumene hydro peroxide, t – butyl peroxide etc., di alkyl peroxide such as di cumyl peroxide, t – butyl cumyl peroxide, di – t – butyl peroxide, tris (t – butyl peroxy) tri azine etc., peroxy ketal such as 1, 1 – di – t – butyl peroxy cyclo hexane, 2, 2 – di (t – butyl peroxy) butane etc., alkyl per ester such as t – butyl peroxy pivalate, t – butyl peroxy – 2 – ethyl hexanoate, t – butyl peroxy iso butylate, di - t – butyl peroxy hexa hydro terephthalate, di - t – butyl peroxy azelate, t – butyl peroxy – 3, 5, 5 – tri methyl hexanoate, t – butyl peroxy acetate, t – butyl peroxy benzoate, di - t – butyl peroxy tri methyl adipate etc., per carbonate such as di iso propyl peroxy di carbonate, di – sec – butyl peroxy di carbonate, t – butyl peroxy di carbonate etc. can be given.

As regards the radical polymerization initiator that can be used at the time of carrying out polymerization by ultraviolet rays, carbonyl compound such as acetophenone, 2, 2 – di methoxy – 2 – phenyl acetophenone, 2, 2 – di ethoxy acetophenone, 4' – iso propyl – 2 – hydroxy – 2 – methyl

propiophenone, 2 – hydroxy – 2 – methyl propiophenone, 4, 4' – bis (di ethyl amino) benzophenone, benzophenone, methyl (o – benzoyl) benzoate, 1 – phenyl 1, 2 – propane dion – 2 – (o – ethoxy carbonyl) oxime, 1 – phenyl – 1, 2 – propane dion – 2 – (o – benzoyl) oxime, benzoin, benzoin methyl ether, benzoin ethyl ether, benzoin iso propyl ether, benzoin iso butyl ether, benzoin octyl ether, benzyl, benzyl di methyl ketal, benzyl di ethyl ketal, di acetyl etc., anthraquinone or thio xanthone derivative such as methyl anthraquinone, chloro anthraquinone, chloro thio xanthone, 2 – methyl thio xanthone, 2 – iso propyl thio xanthone etc., sulfur compounds such as di phenyl di sulfide, di thio carbamate etc. can be given.

The usage quantity of radical polymerization initiator differs with the type of radical polymerization initiator, type and composition ratio of monomer, however, it should be within the range from 0.001 ~ 20 mole %, desirably 0.01 ~ 10 mole % with respect to monomer component. If the usage quantity of radical polymerization initiator is less than 0.001 mole %, polymerization requires long time whereas if it is more than 20 mole %, then it is not only uneconomical, but bubbles are generated during the polymerization and molecular weight of the hardened material obtained by polymerization remarkably reduces which is not desired.

The polymerizable unimer of the present invention can be polymerized and hardened as it is or it can be preparatory polymerized and then polymerization and hardening is carried out based on which viscosity can be regulated and shrinkage ratio at the time of polymerization can be reduced.

[Practical examples]

The present invention has been explained below in further details with the help of practical examples and reference examples. However, the present invention is not restricted only to these practical examples.

(Practical example 1)

50.0 g of 4, 4' – bis (2 – hydroxy ethyl thio) phenyl sulfide that is divalent alcohol and 300 ml of benzene were taken in a separable flask (500 ml) having 3 openings equipped with a thermometer, Dimroth condenser, stirrer and moisture supplier (?) and it was dissolved and acrylic acid, p – toluene sulfonic acid and hydroquinone were added to this solution and reflux was carried out by heating for 10 hours. The obtained solution was extracted by adding aqueous solution of 1 N sodium hydroxide after which it was repeatedly washed with water till it became neutral. The organic solution obtained after washing was dried by adding anhydrous magnesium sulfate and solvent was removed under reduced pressure when the crude product was obtained. This crude product was refined by using silica gel column chromatography (effluent (?) solvent: hexane / ethyl acetate = 7/3) when the aimed 4, 4' – bis [2 - acryloyloxy ethyl thio] phenyl sulfide was obtained.

The obtained 4, 4' – bis [2 - acryloyloxy ethyl thio] phenyl sulfide was colorless transparent

oily liquid at normal temperature (boiling point: > 200°C gelation). Elemental analysis of this compound are as follows.

Refractive index: $N_D = 1.6291$

(Shimazu Seisakujo make ML model refractometer, measurement temperature: 25°C)

Elemental analysis $C_{22}H_{22}O_4S_2 = 446.59$

Calculated values C: 59.17 %, H: 4.97 %, S: 21.36 %

Experimental values C: 59.02 %, H: 5.13 %, S: 21.81 %

Moreover, IR spectrum chart of the obtained 4, 4' – bis [2 - acryloyloxy ethyl thio) phenyl sulfide has been shown in figure 1 and 1H – NMR spectrum chart has been shown in figure 2. IR spectrum was measured by using Hitachi 270 – 50 photospectrometer and 1H – NMR spectrum was measured by using Nippon Denshi JNM – PMX 60 (60 MHz) NMR measuring device using TMS (tetra methyl silane) as internal standard.

(Practical example 2)

50.0 g of 4, 4' – bis (ω - hydroxy polyoxy propyl thio) phenyl sulfide (in formula (II), $R_2 = -CH_3$ and $n = m = 2$) that is divalent alcohol and 300 ml of methylene chloride were taken in a separable flask (500 ml) having 3 openings equipped with a thermometer, Dimroth condenser, stirrer and moisture supplier (?) and it was dissolved and 940 mg of tri ethylamine was added this solution it was cooled to 0 ~ 5°C. Next, 22.8 g of methacrylic acid chloride refined beforehand by distillation was gradually dropped by a dropping funnel simultaneously while maintaining the temperature between 0 ~ 5°C. After the completion of dropping, stirring was continued at room temperature for approximately 3 hours after which this solution was extracted by adding aqueous solution of 1 N sodium hydroxide and then it was repeatedly washed with water till it became neutral. The organic solution obtained after washing was dried by adding anhydrous magnesium sulfate and solvent was removed under reduced pressure when the crude product was obtained. This crude product was refined by using silica gel column chromatography (effluent (?) solvent: hexane / ethyl acetate = 7/3) when the aimed colorless transparent liquid 4, 4' – bis (ω - methacryloyloxy propyl thio) phenyl sulfide (in formula (I), $R_1 = -CH_3$, $R_2 = -CH_2$, $n = m = 2$) was obtained.

The refractive index, elemental analysis and spectrum data of the obtained aimed compound are as follows.

Refractive index: $N_D = 1.5960$

(Shimazu Seisakujo make ML model refractometer, measurement temperature: 25°C)

Elemental analysis $C_{22}H_{42}O_8S_4 = 618.86$

Calculated values C: 62.11 %, H: 6.84 %, S: 15.54 %

Experimental values C: 62.02 %, H: 6.75 %, S: 15.83 %

IR: ν cm⁻¹

1740 (ester carbonyl)

1590 and 1490 (phenyl nucleus)

1180 (ether), 810 (phenyl out-of-plane bending angle ?)

¹H - NMR: δ (CDCl₃)

7.23 (8 H, s), 6.08 (2 H, m)

5.64 (2 H, m), 4.6 ~ 3.8 (10 H, m)

3.28 (2 H, m)

1.98 (6 H, d, J = 2 Hz)

1.25 (6 H, d, J = 7 Hz)

1.25 (6 H, d, J = 7 Hz)

(Practical example 3)

4, 4' - bis (ω - hydroxy polyoxy ethyl thio) phenyl sulfide (in formula (II), R₂ = H and n = m = 2) that is divalent alcohol was used instead of 4, 4' - bis (2 - hydroxy ethyl thio) phenyl sulfide. Other than this change, method similar to practical example 1 was used and the aimed 4, 4' - bis (ω - methacryloyloxy polyoxy ethyl thio) phenyl sulfide (in formula (I), R₁ = - H, R₂ = - H, n = m = 2) was obtained.

The refractive index, elemental analysis and spectrum data of the obtained aimed compound are as follows.

Refractive index: N_D = 1.6075

(Shimazu Seisakujo make 3L model refractometer, measurement temperature: 25°C)

Elemental analysis C₂₄H₃₀O₆S₃ = 534.70

Calculated values C: 58.40 %, H: 5.65 %, S: 17.99 %

Experimental values C: 58.43 %, H: 5.51 %, S: 17.65 %

IR: ν cm⁻¹

1730 (ester carbonyl)

1590 and 1480 (phenyl nucleus)

1160 (ether), 810 (phenyl out-of-plane bending angle ?)

¹H - NMR: δ (CDCl₃)

7.28 (8 H. s)

6.44 (2 H. dd. J = 2. 17 Hz)

6.13 (2 H. dd. J = 11. 17 Hz)

5.80 (2 H. dd. J = 2. 11 Hz)

4.53 (4 H. t. J = 6 Hz)

4.20 (4 H. t. J = 6 Hz)

4.13 (4 H. t. J = 6 Hz)

3.34 (4 H. t. J = 6 Hz)

(Practical example 4)

4, 4' – bis (ω - hydroxy polyoxy ethyl thio) phenyl sulfide (in formula (II), $R_2 = H$ and $n = m = 2$) that is divalent alcohol was used instead of 4, 4' – bis (ω - hydroxy polyoxy propyl thio) phenyl sulfide (in formula (II), $R_2 = -CH_3$ and $n = m = 2$). Other than this change, method similar to practical example 2 was used and the aimed 4, 4' – bis (ω - methacryloyloxy polyoxy ethyl thio) phenyl sulfide (in formula (I), $R_1 = -CH_2$, $R_2 = -H$, $n = m = 4$) was obtained.

The refractive index, elemental analysis and spectrum data of the obtained aimed compound are as follows.

Refractive index: $N_D = 1.5871$

(Shimazu Seisakujo make 3L model refractometer, measurement temperature: 25°C)

Elemental analysis $C_{30}H_{50}O_{10}S_3 = 738.97$

Calculated values C: 58.51 %, H: 6.82 %, S: 13.02 %

Experimental values C: 58.44 %, H: 6.59 %, S: 13.33 %

IR: ν cm⁻¹

1735 (ester carbonyl)

1580 and 1490 (phenyl nucleus)

1160 (ether), 810 (phenyl out-of-plane bending angle ?)

1H – NMR: δ (CDCl₃)

7.25 (8 H. s) . 6.10 (2 H. m)
5.62 (2 H. m)
4.51 (4 H. t . J = 6 Hz)
4.18 (2.4 H. br t . J = 6 Hz)
3.31 (4 H. t . J = 6 Hz)
1.99 (6 H. d . J = 2 Hz)

(Reference example 1)

20 mg of 2, 2' – azo bis (2, 4 – di methyl valeronitrile) taken as radical polymerization initiator was dissolved in 20 g of 4, 4' – bis (2 – acryloyloxy ethyl thio) phenyl sulfide obtained in practical example 1 and it was injected in glass cell of 0.2 cm x 20 cm x 20 cm and glass mold of 0.2 mm x 75 mm ϕ respectively and heating was carried out at 35 $^{\circ}$ C under nitrogen current for 10 hours after which it was heated up to 80 $^{\circ}$ C with the rate of rise of temperature of 10 $^{\circ}$ C/hr and the mold was released and hardening was further carried out by heating at 100 $^{\circ}$ C for 1 hour.

The obtained hardened material was colorless transparent and it possessed properties given below.

Tg 150 $^{\circ}$ C

(Orientech make Leopyron DDV – II – EP model)

Moisture absorbance 0.2 % (ASTM D-570 – 59T)

(soaking at 23 $^{\circ}$ C, 24 h)

Refractive index $N_D^{2\alpha} = 1.656$

(Shimazu Seisakujo make ML model refractometer, measurement temperature: 25 $^{\circ}$ C)

(Reference example 2)

50 mg of 2, 2' – azo bis (2, 4 – di methyl valeronitrile) taken as radical polymerization initiator was dissolved in 50 mg of hardening composite formed from 35 g of 4, 4' – bis (ω - methacryloyloxy ethyl thio) phenyl sulfide (in formula (I), $R_1 = -H$, $R_2 = -H$, $n = m = 2$) obtained in practical example 3 and 15 g of methyl methacrylate and it was injected in glass mold of 5 cm x 5 cm x 0.3 cm and glass cell of 0.2 mm x 50 mm x 20 mm respectively and heating was carried out at 35 $^{\circ}$ C under nitrogen current for 10 hours after which it was heated up to 80 $^{\circ}$ C with the rate of rise of temperature of 10 $^{\circ}$ C/hr and the mold was released and hardening was further carried out by heating at 100 $^{\circ}$ C for 1 hour.

The obtained hardened material was colorless transparent and it possessed properties given below.

Tg 135 $^{\circ}$ C

(Orientech make Leopypron DDV - II - EP model)

Moisture absorbance 0.22 % (ASTM D-570 - 59T)

(soaking at 23°C, 24 h)

Refractive index $N_D^{2\alpha} = 1.588$

(Shimazu Seisakujo make ML model refractometer, measurement temperature: 25°C)

4. Brief explanation of figures

Figure 1 is IR spectrum chart of the compound obtained in practical example 1. Figure 2 is the ^1H - NMR spectrum chart of the compound obtained in practical example 1.

Patent Applicant

Showa Denko Kabushiki Kaisha

(Showa Electrical Co. Ltd.)

Representative Patent Attorney Kikuchi Seiichi

Figure 1

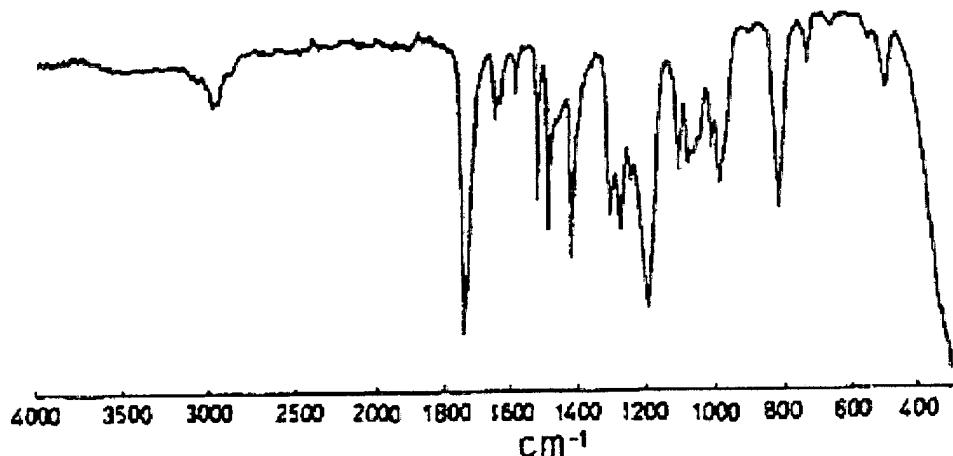
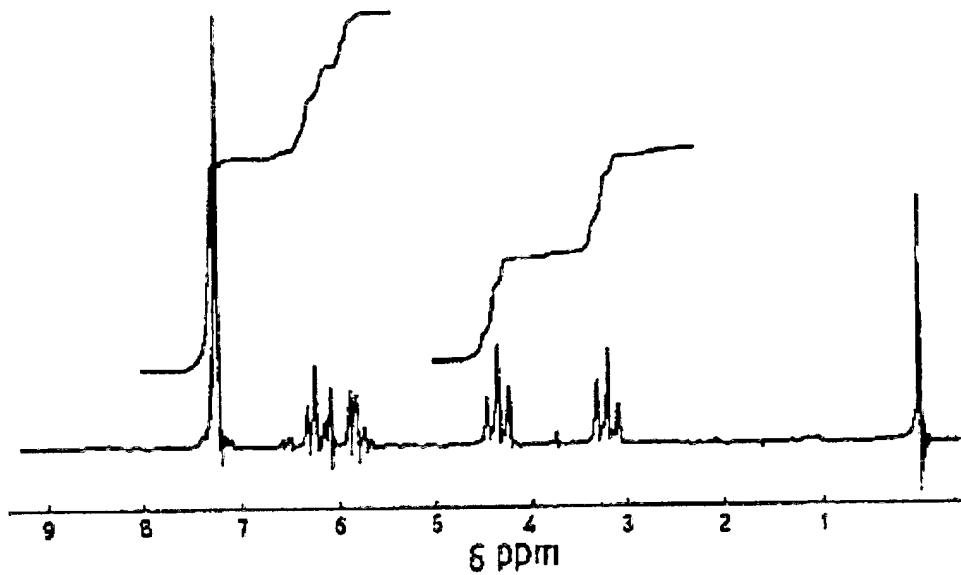


Figure 2

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(51) Int. Cl.⁵ Discrimination Internal Arrangement

Number	Number		
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	MMV	A	8620 - 4J
(72) Inventor			
Arakawa Tsukasa			
Kanagawa ken, Kawasaki shi, Kawasaki ku,			
Chidori cho, 3 - 2.			
Showa Denko Kabushiki Kaisha Kawasaki			
Juki Kenkyusho Nai			
(Internal to Kawasaki Resin Research Center			
of Showa Electrical Co. Ltd.)			

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